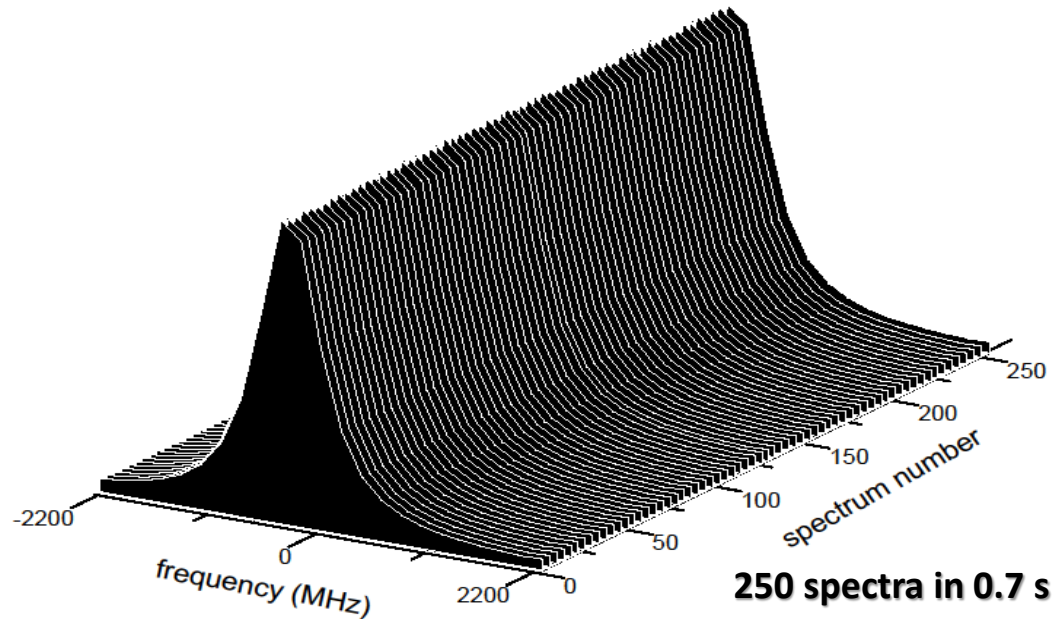


Sensor Development: Metrology Tools for Climate Science



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National Institute of Standards and Technology • U.S. Department of Commerce

Our approach

- Both *in situ* monitoring and remote sensing require cutting-edge spectroscopic measurements
- We develop novel techniques which allow for enhanced:
 - Spectral coverage
 - Portability
 - Sensitivity
 - Accuracy
 - Speed
 - Selectivity

Outline

- **A series of new spectroscopic techniques**
 - **They offer a range of complexity, speed, and sensitivity**
- **Specifically I will discuss:**
 - **Photoacoustic sensor**
 - **Ultrasensitive cavity ring-down instruments**
 - **Multiplexed detection with optical frequency combs**

NIST photoacoustic sensor

- Designed for routine monitoring of CO₂ concentrations
- Will be placed on the top of the Admin. Building at NIST (101).

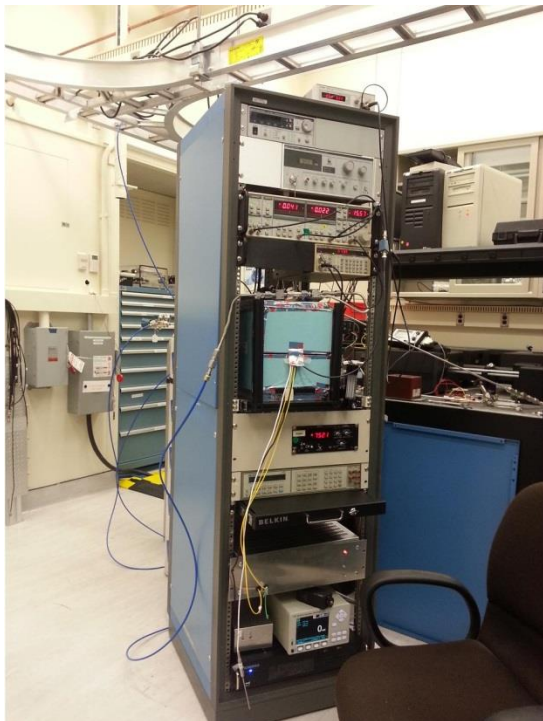
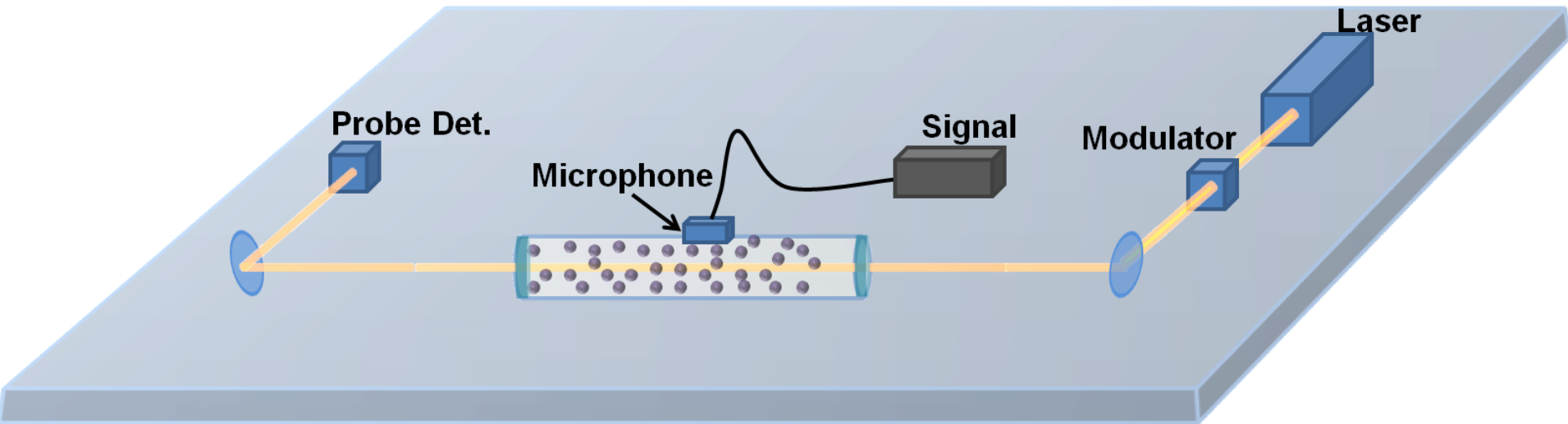


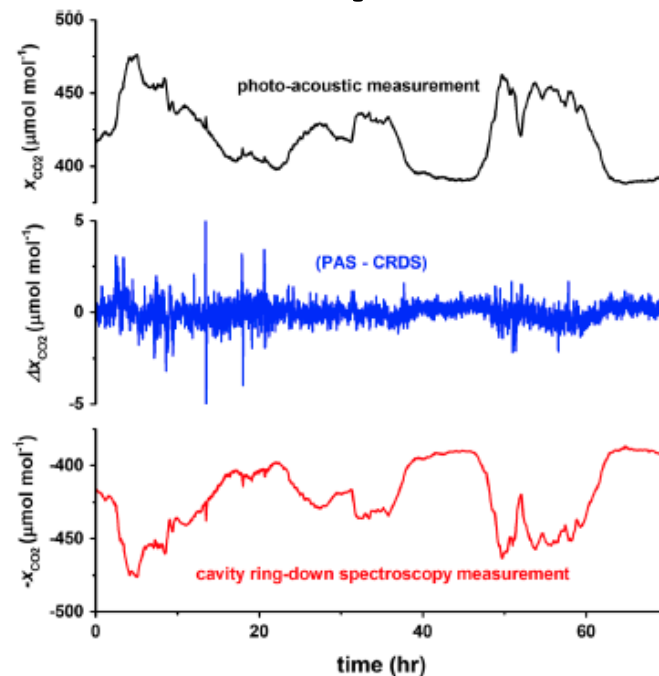
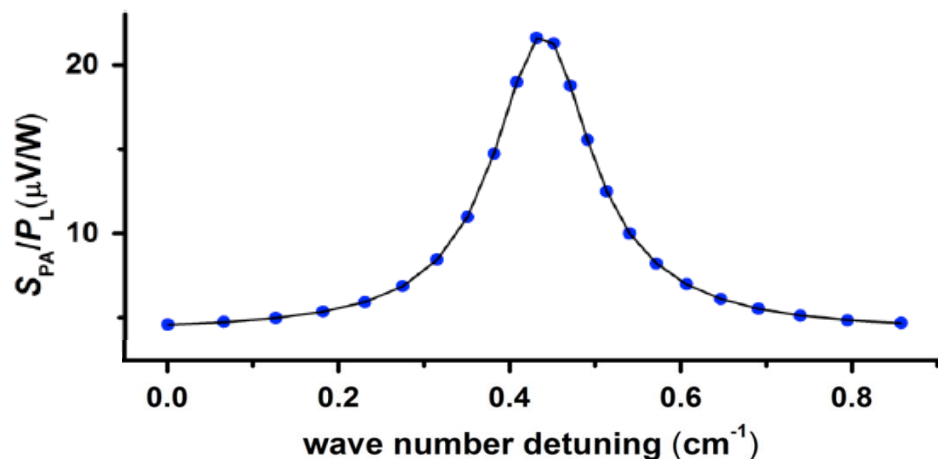
Figure from HDR Architecture, Inc.

Photoacoustic spectroscopy (PAS)



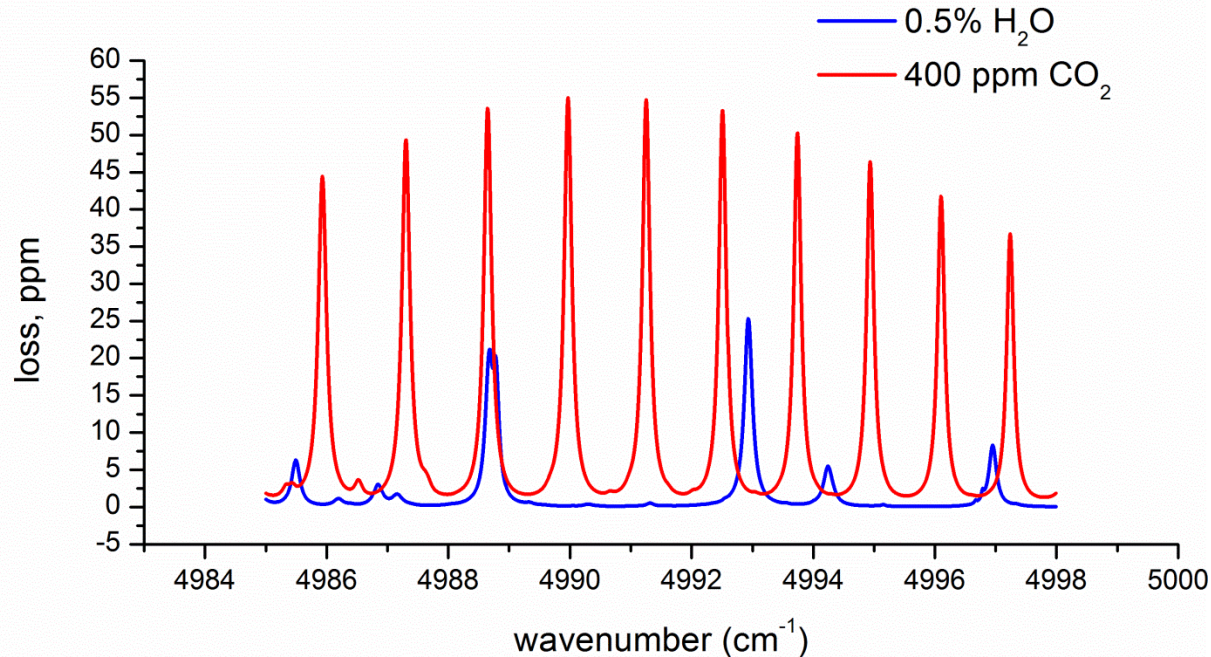
- Zero background
- Optically broadband
- Relatively easy to implement
- High sensitivity
 - signal scales with laser power
- Wide dynamic range

Photoacoustic spectrometer at 1.6 μm



- Allows for routine, automated measurements of ambient CO₂
- Uncertainty of only 0.8 ppm (0.2%)

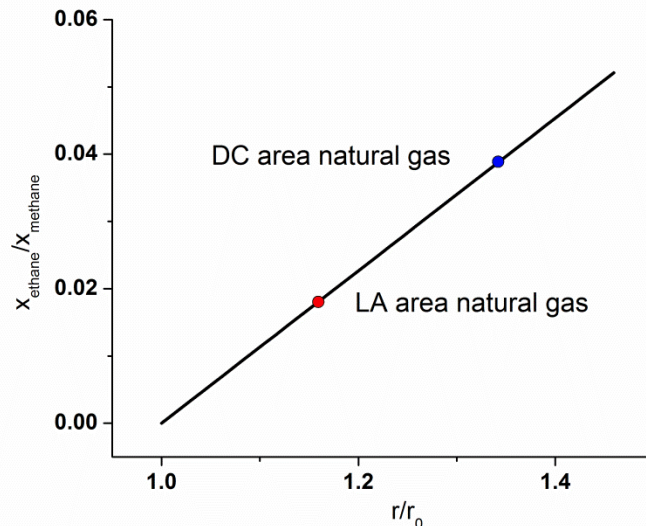
New 2 μm sensor



- **Moved to 2 μm to probe stronger CO_2 transitions**
 - Able to utilize recently developed laser technology
- **Signal-to-noise ratios of $\sim 14,000:1$ for CO_2 at ambient levels**
- **Allows for simultaneous humidity measurements with the same laser**

Future directions for the PAS instruments

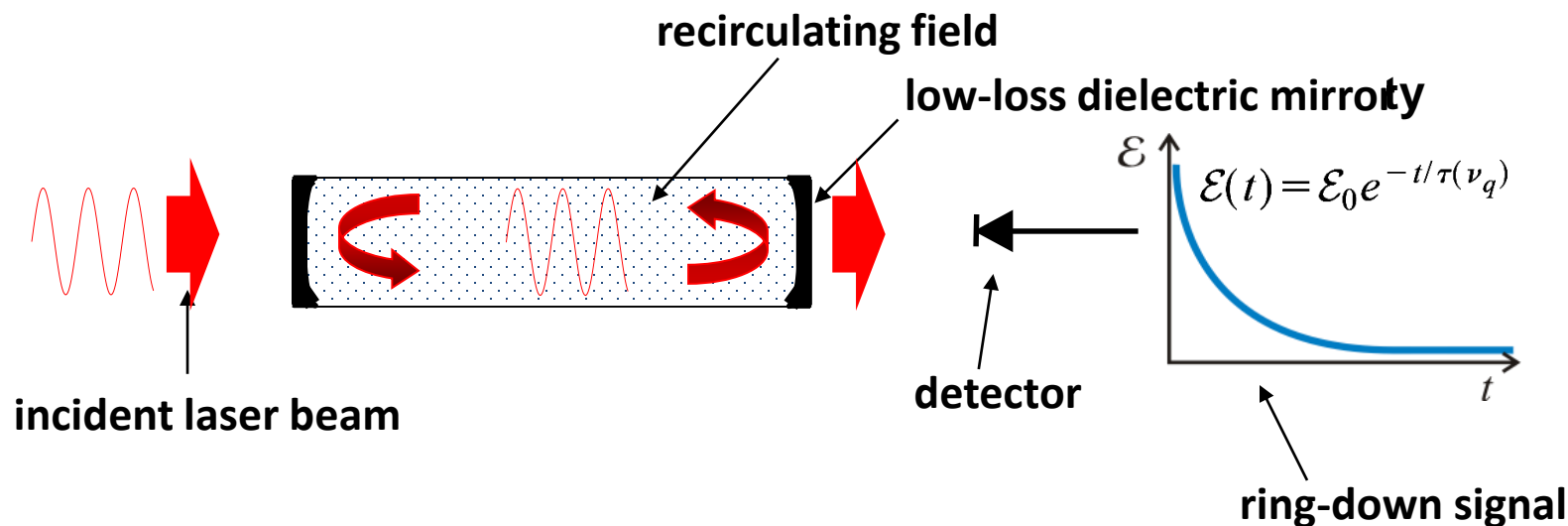
- **Dual species PAS instrument**
 - Use fiber switches to probe numerous species simultaneously
 - Potential targets include CO_2 , H_2O , NH_3 , and CH_4 (and their isotopologues)
- **Mid-infrared PAS**
 - Would allow us to probe CH_4 and C_2H_6 simultaneously
 - Allows for source attribution of CH_4 emissions



Outline

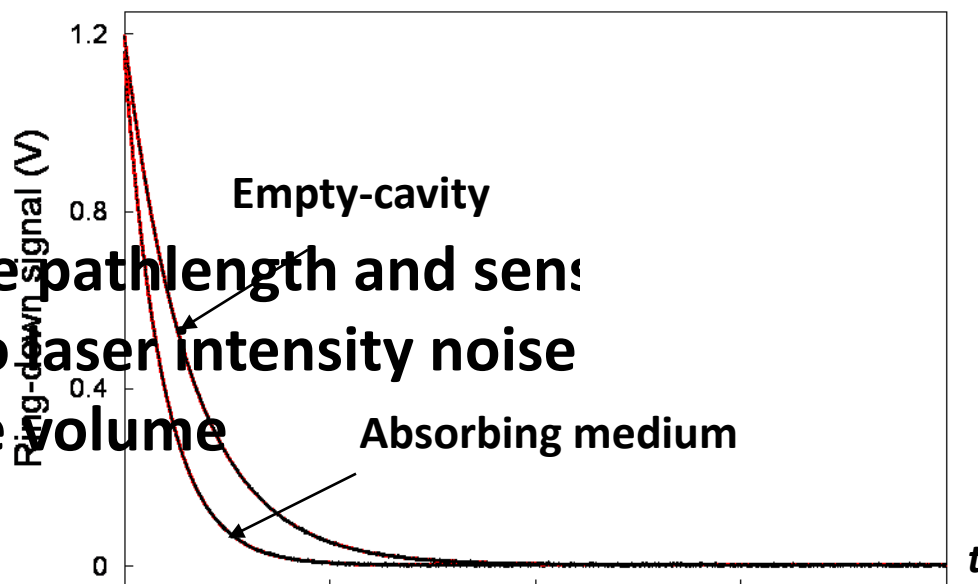
- **A series of new spectroscopic techniques**
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Cavity ring-down spectroscopy (CRDS)



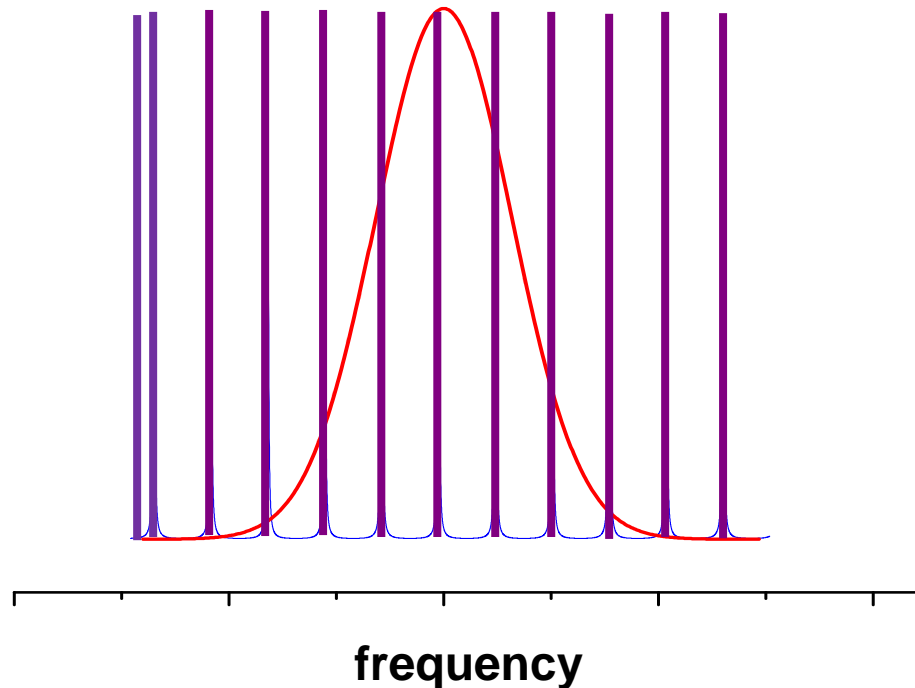
Advantages:

- High effective pathlength and sensitivity
- Insensitive to laser intensity noise
- Small sample volume



The problem

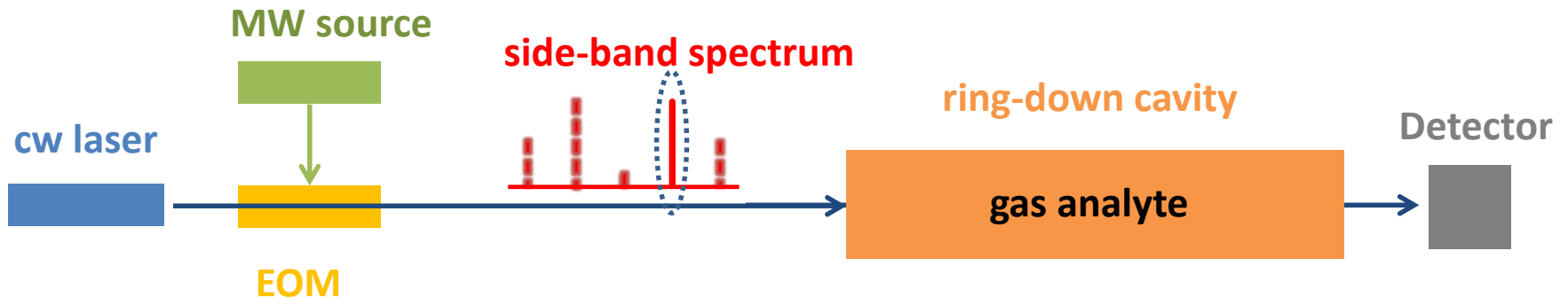
- To record a spectrum you need to tune the laser frequency
- This generally requires thermal or mechanical tuning
 - This is usually non-linear and very slow
- Things are even more difficult with cavity-enhanced spectroscopy (discrete frequencies)



Frequency-agile, rapid scanning (FARS) spectroscopy

Method:

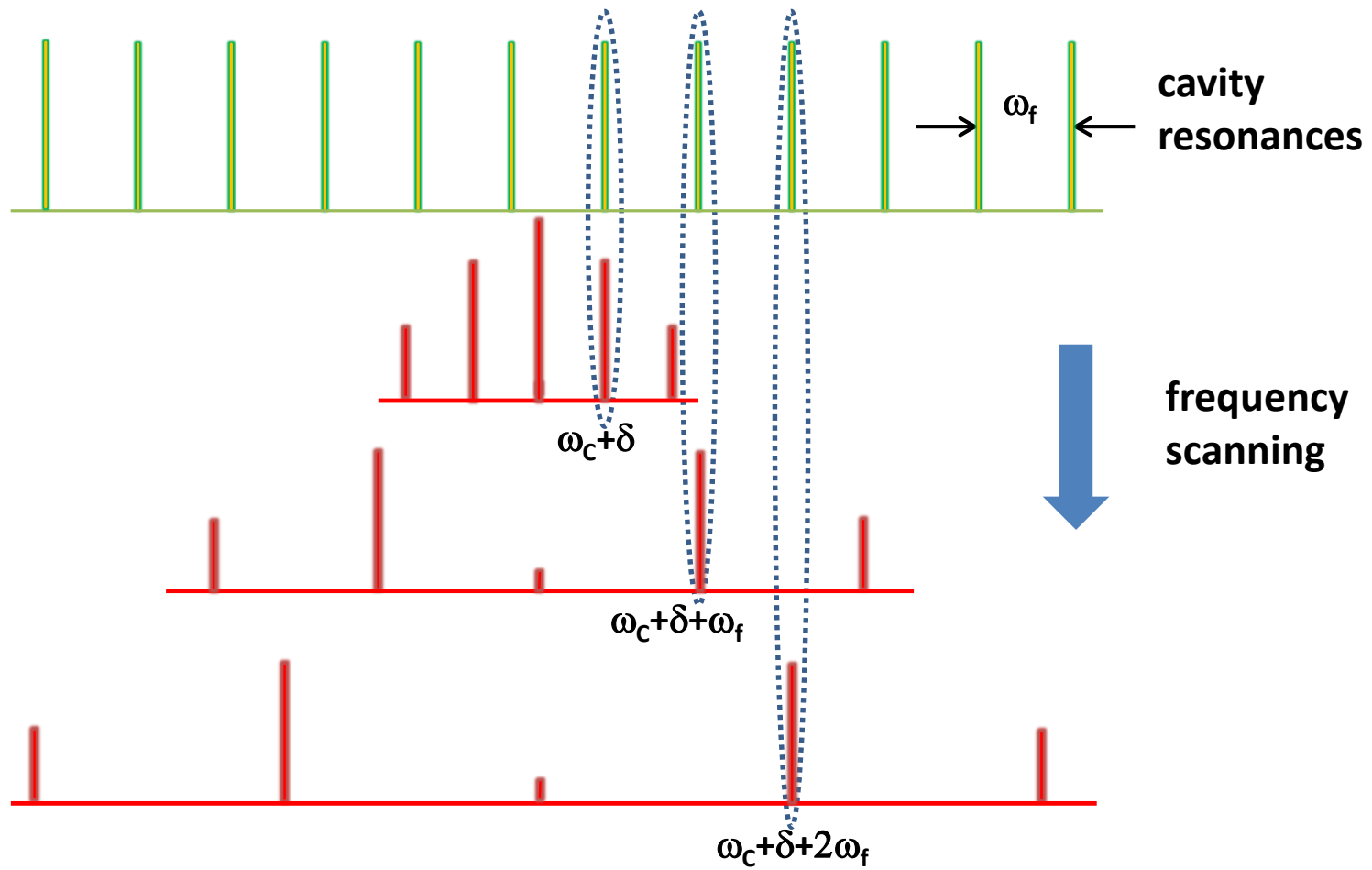
- Use waveguide electro-optic phase-modulator (EOM) to generate tunable sidebands
- Drive PM with a rapidly-switchable microwave (MW) source
- Fix carrier and use ring-down cavity to filter out all but one selected side band



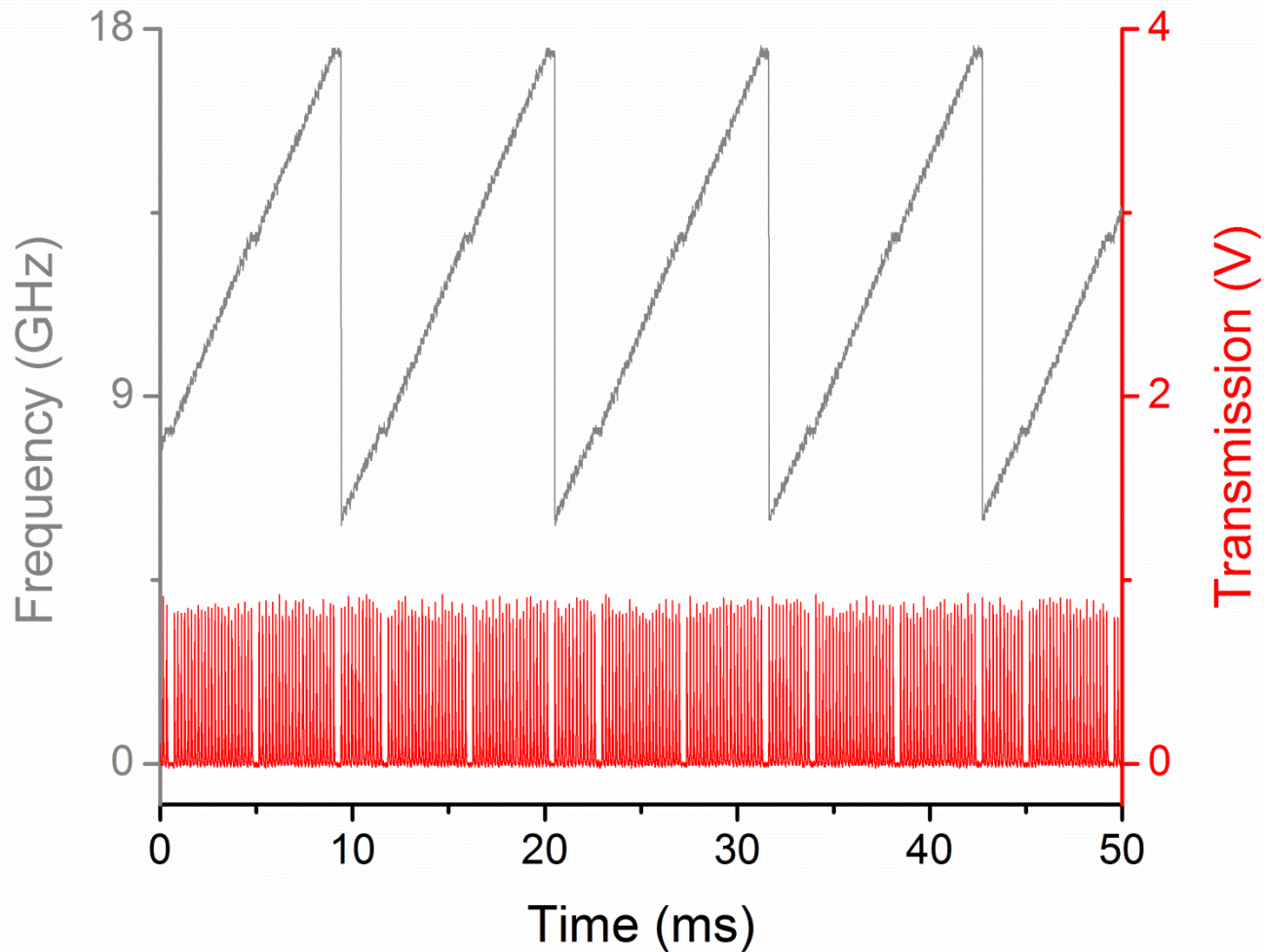
Advantages:

- Overcomes slow mechanical and thermal scanning
- Links optical detuning axis link to radio-frequency (RF) standards
- Wide frequency tuning range ($> 130 \text{ GHz} = 4.3 \text{ cm}^{-1}$)

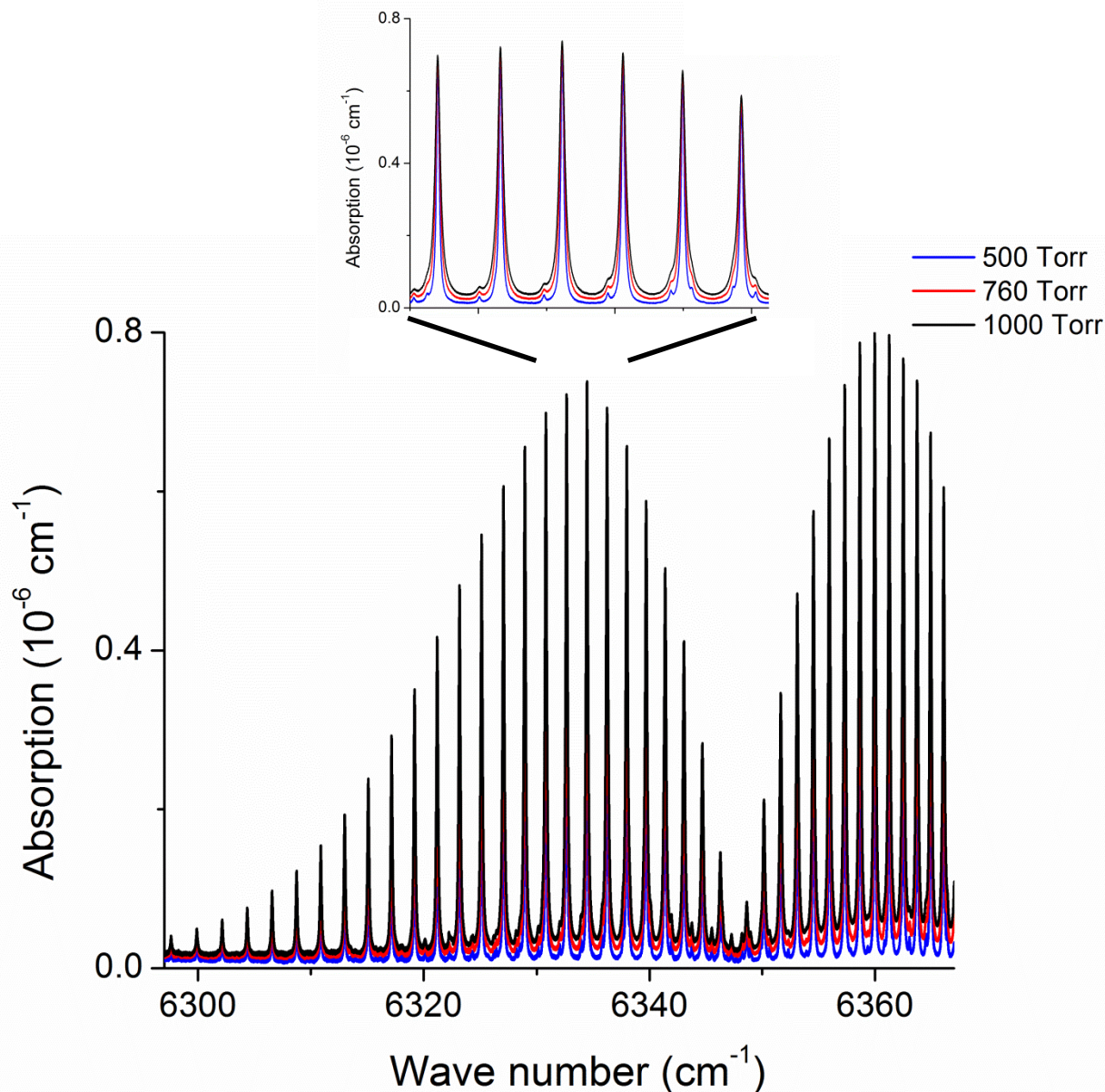
FARS operating principle



Very high acquisition rates: scanning



Spectra of entire absorption bands



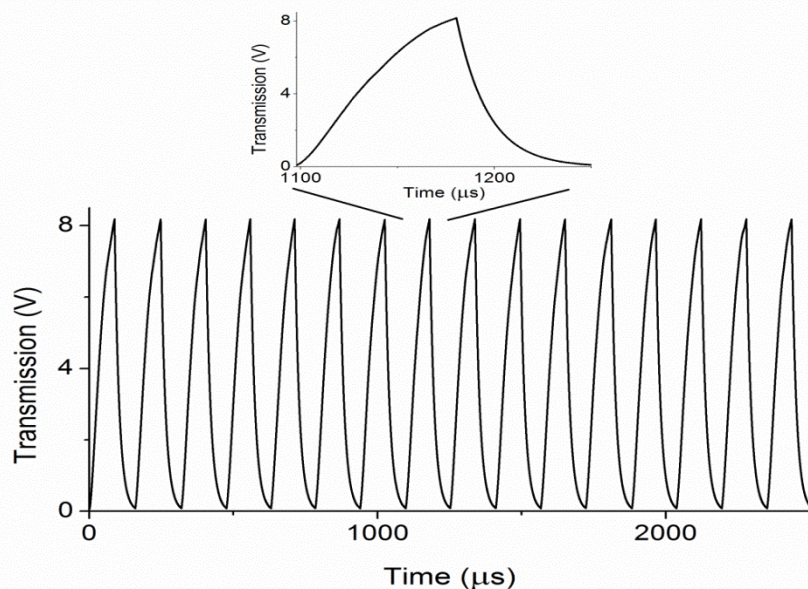
**2 THz wide spectra
recorded in 45 minutes**

425 ppm of CO₂ in air

**ECDL grating moved
every 12 GHz**

**Each point is the
average of 100 RDs**

Lower finesse = faster rates

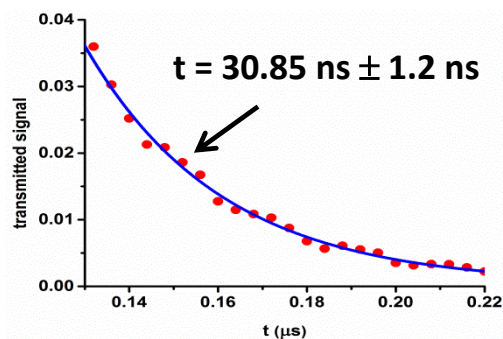
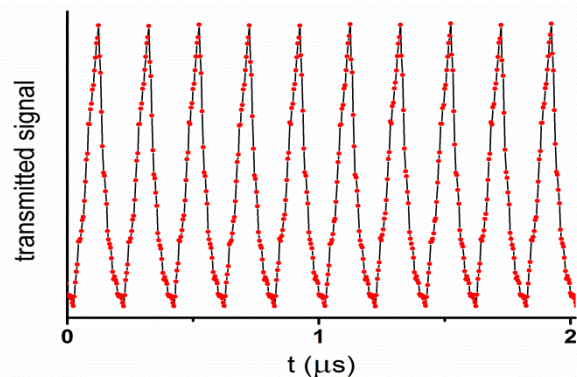


Finesse = 20,000

RD acq. rate = 8 kHz

$\sigma_\tau/\tau = 0.008 \%$

NEA = $1.7 \times 10^{-12} \text{ cm}^{-1} \text{ Hz}^{-1/2}$



Finesse = 60

RD acq. rate = 5 MHz

$\sigma_\tau/\tau = 4 \%$

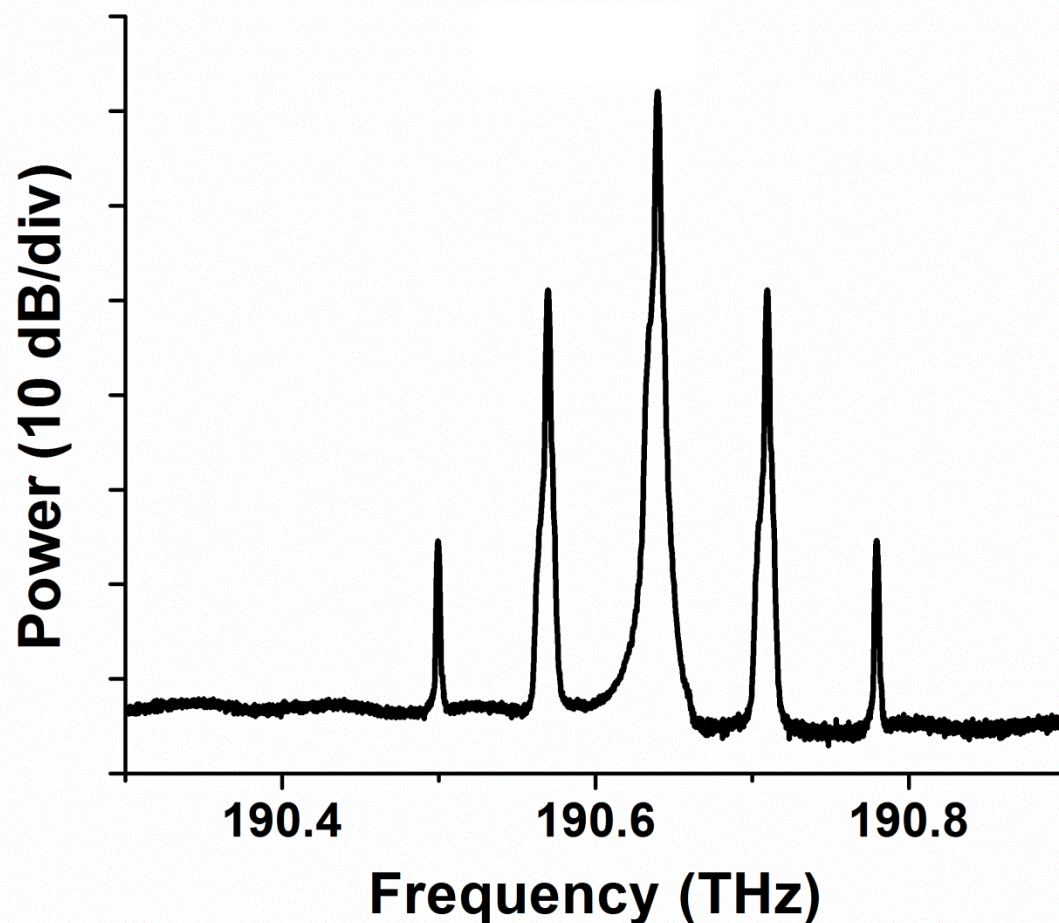
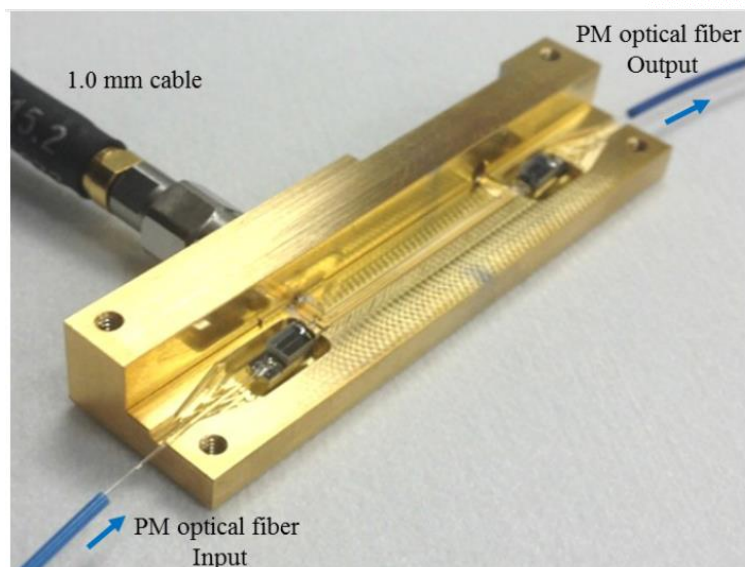
NEA = $1.9 \times 10^{-8} \text{ cm}^{-1} \text{ Hz}^{-1/2}$

What if I want to scan faster?

- **Entirely limited by the slow grating tuning**
- **So use higher bandwidth EOMs to reduce the number of grating steps**

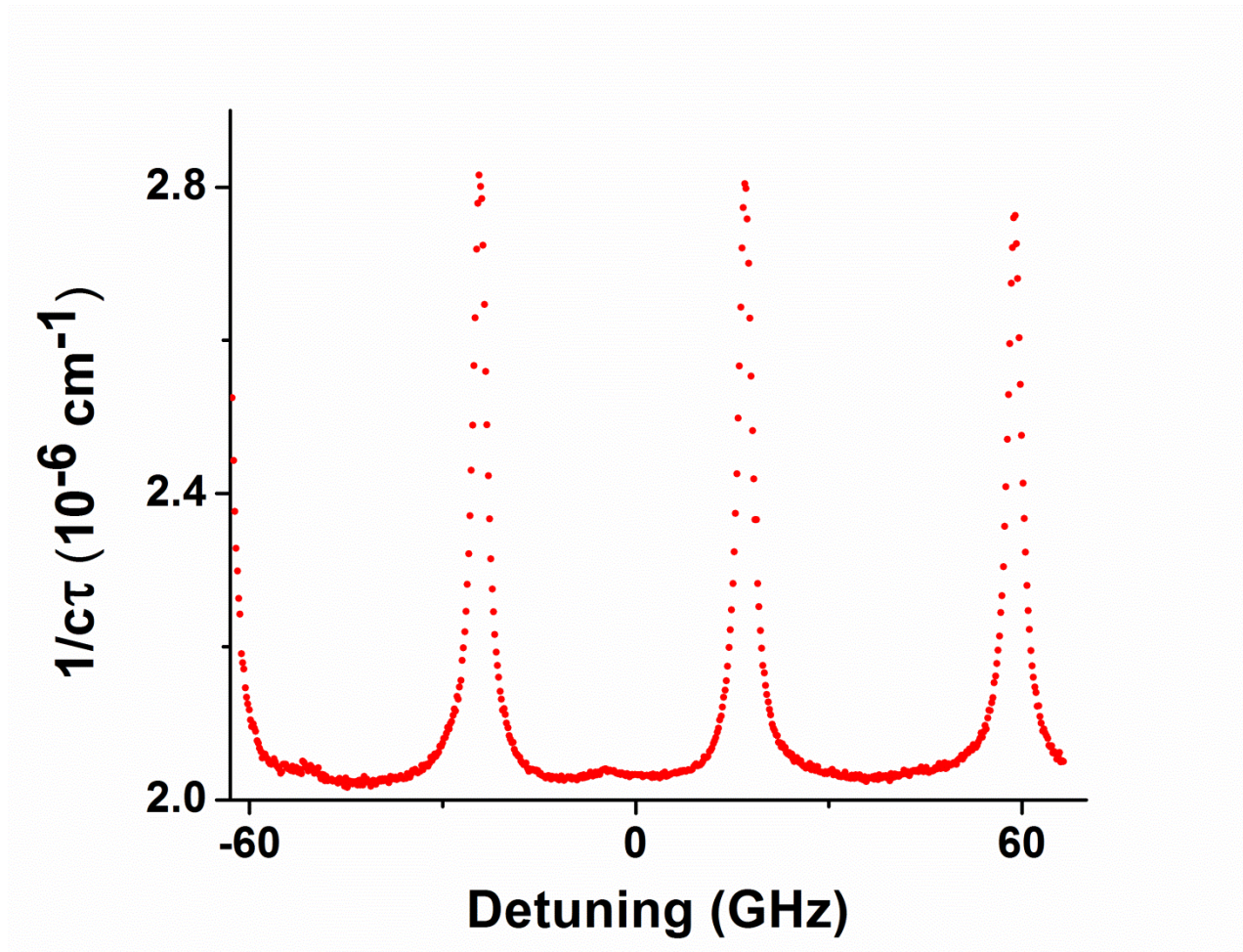
Even faster rates

Use recently developed W-band modulators (bandwidth up to 300 GHz!)

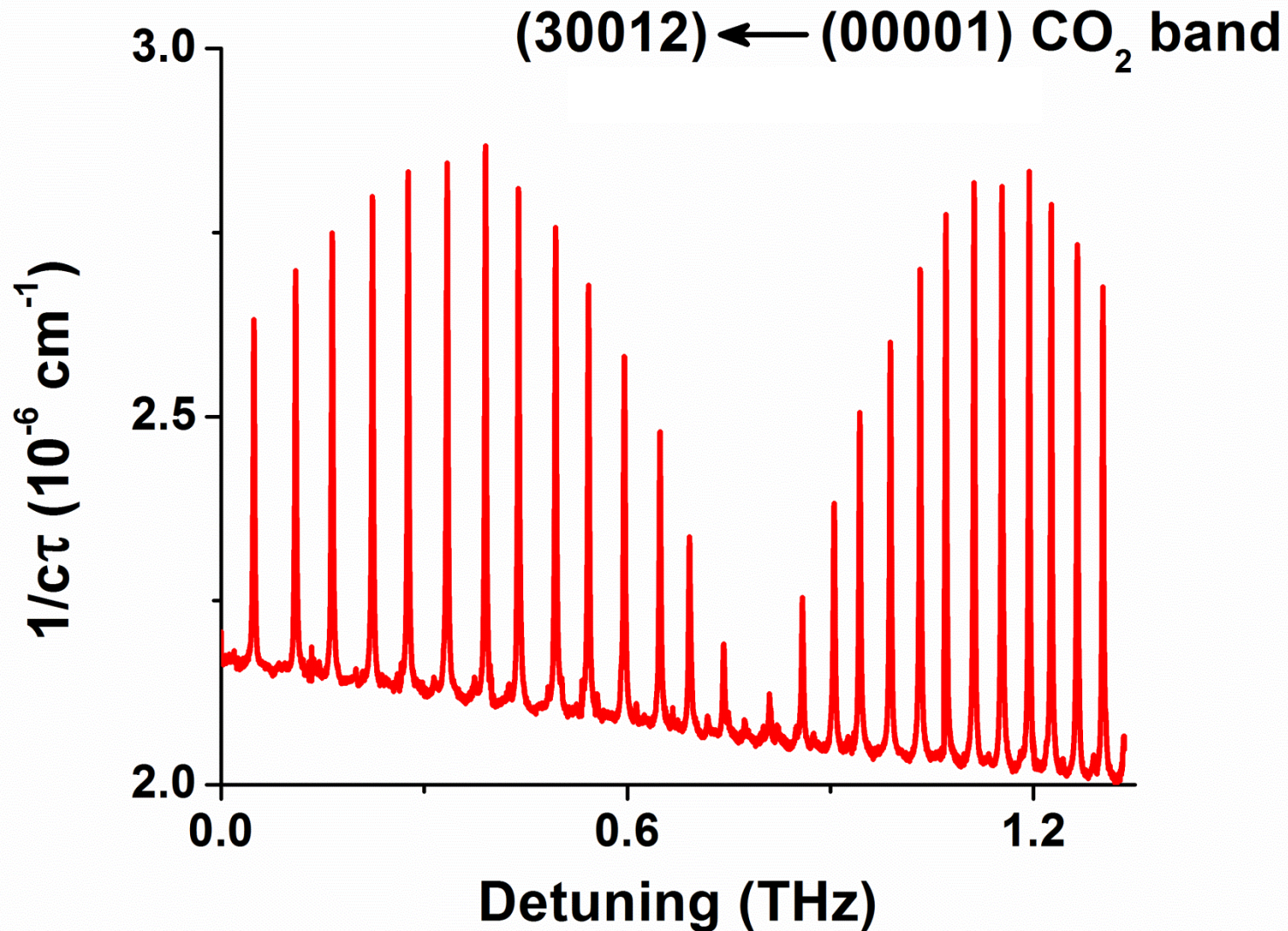


Even faster rates: single grating position

Allows for a 130 GHz (4.3 cm^{-1}) to be recorded in 3 s.



Even faster rates: grating tuning




What if I want higher sensitivity?

- To reduce $1/f$ noise, we want to make the measurement away from DC
- Also need to rapidly compare ring-down time constants at different wavelengths

Improving the sensitivity: Heterodyne measurements

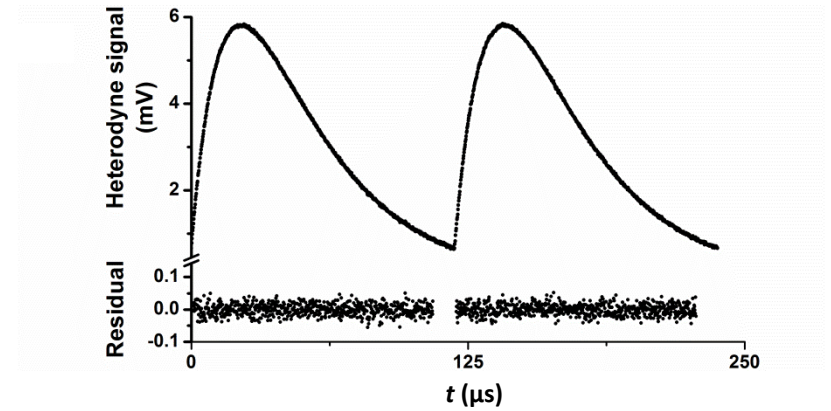
- To do this we adapted the approach of Ye and Hall

[

APD 
High Finesse Cavity

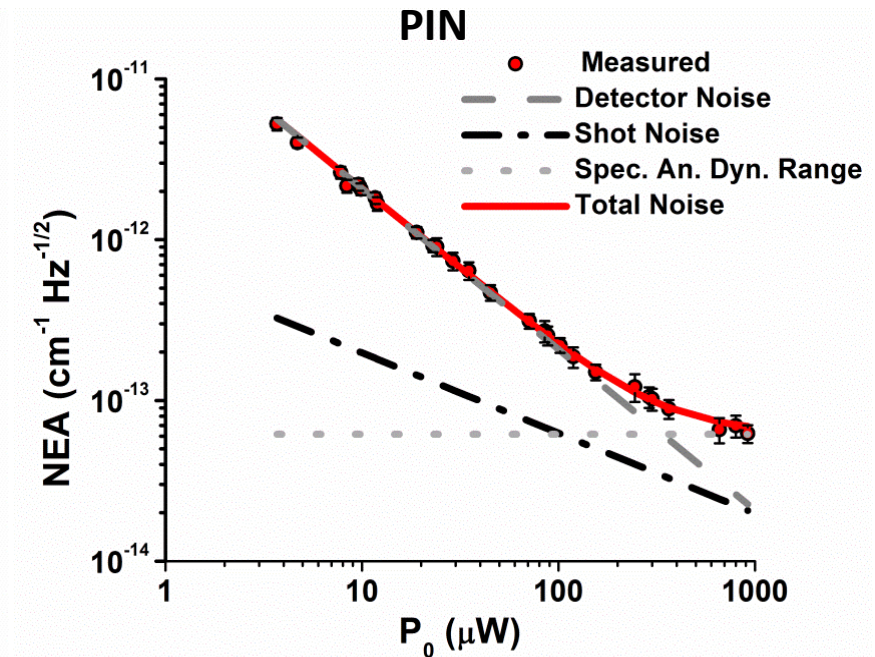
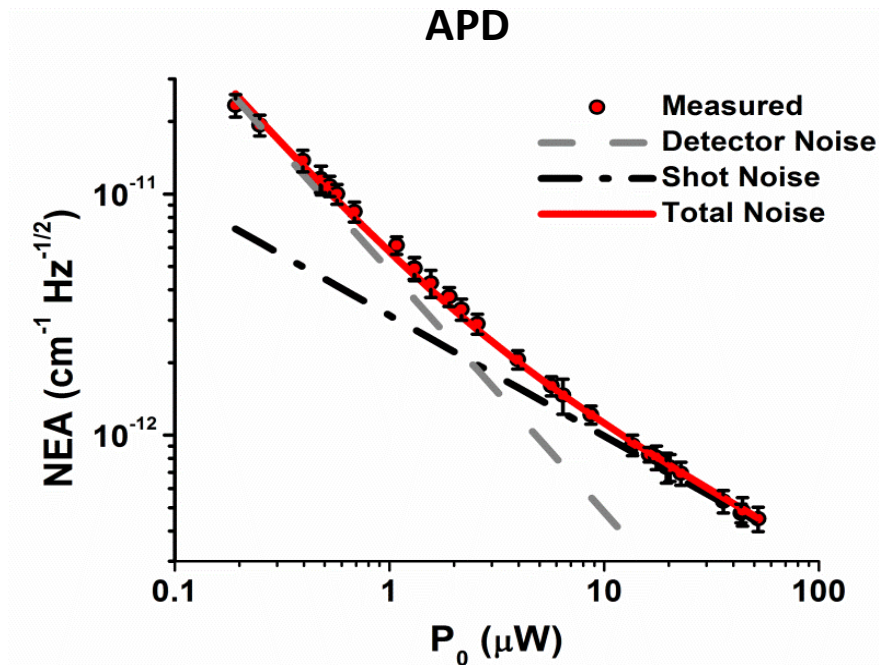
Heterodyne measurements: Our approach

- Replace their two AOMs with a single EOM
- This reduces the complexity and enables rapid scanning



Heterodyne measurements: Reaching the quantum-noise limit

- Utilized both a traditional InGaAs detector and an APD
- Able to reach the quantum-noise limit with the APD
- The traditional InGaAs allows for an NEA of $6\text{E-}14 \text{ cm}^{-1} \text{ Hz}^{-1/2}$



Most sensitive spectrometers

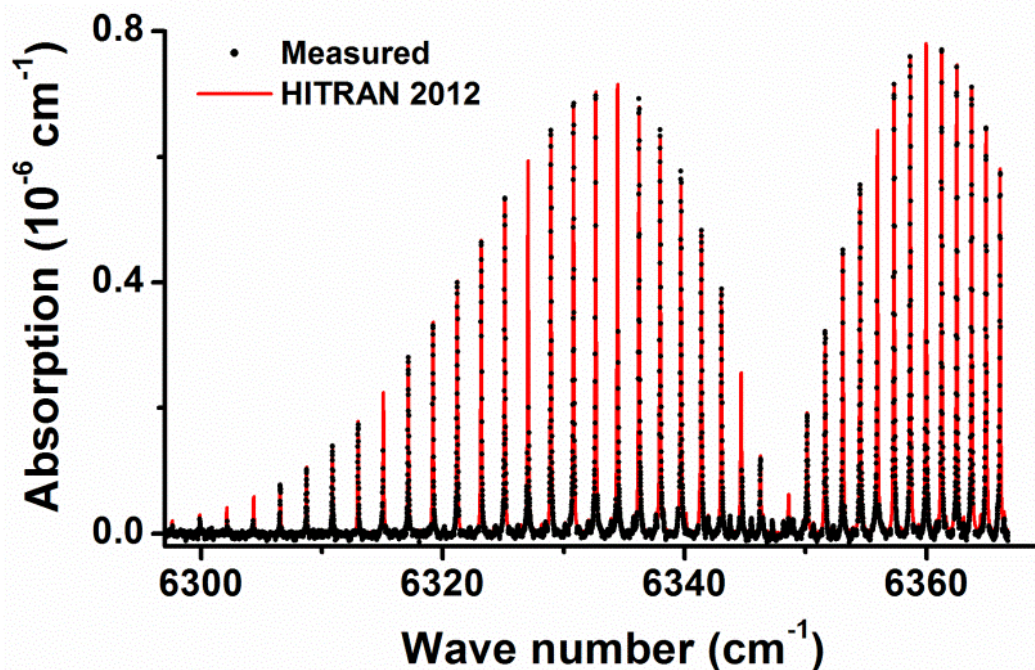
Technique	Ref.	Sensitivity ($\text{cm}^{-1} \text{ Hz}^{-1/2}$)	Laser	Tuning range (nm)
NICE-OHMS	Ye et al.	<u>1E-14</u>	cw-Nd:YAG	0.1
HD-CRDS	Long et al.	<u>6E-14</u>	ECDL	60
HD-CRDS	Ye and Hall	3E-13	cw-Yb:YAG	~0.5
CRDS	Spence et al.	1E-12	cw-Nd:YAG	0.14
FARS-CRDS	Long et al.	2E-12	ECDL	60
NICE-OHMS	Ehlers et al.	6E-12	Fiber laser	1

D. A. Long, et al., Appl. Phys. B, Under Review (2014)

D. A. Long et al., Appl. Phys. B, (2013)

Heterodyne measurements: Rapid scanning

- Able to record a 26 GHz-wide spectrum in 17 ms with 50 GS/s AWG
- Observed a weak CO₂ hot band transition ($3.5\text{E-}25 \text{ cm molec.}^{-1}$) in an air sample



Outline

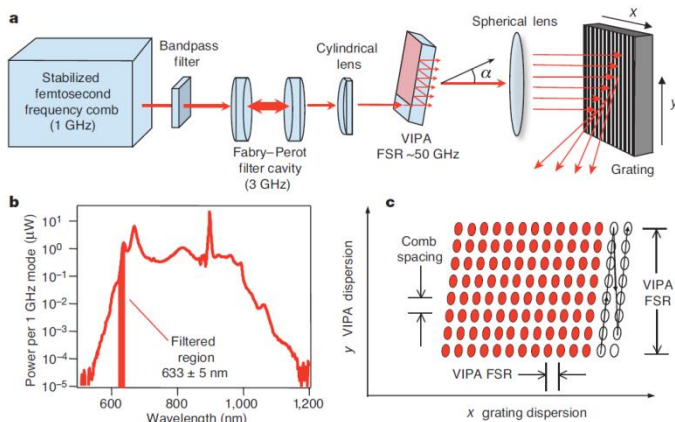
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What if I don't want to scan?

- Then multiplex!

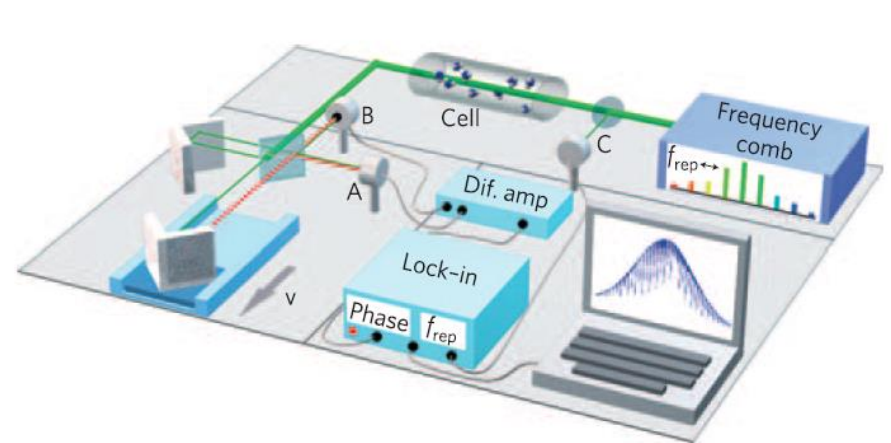
Multiplexed measurements: OFCs

OFCs have been used with a variety of detection schemes for spectroscopic measurements



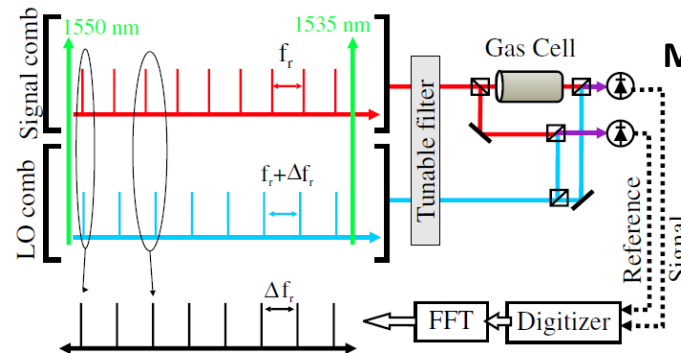
Dispersive (VIPA)

Diddams, et al., Nature (2007)



Fourier Transform

Mandon, et al., Nat. Photonics (2009)



Multiheterodyne

Coddington, et al., Phys. Rev. Lett. (2008)

Mode-locked femtosecond OFCs



Advantages:

- Wide bandwidth (octave-spanning)
- Can be self-referenced (absolute freq. axis)

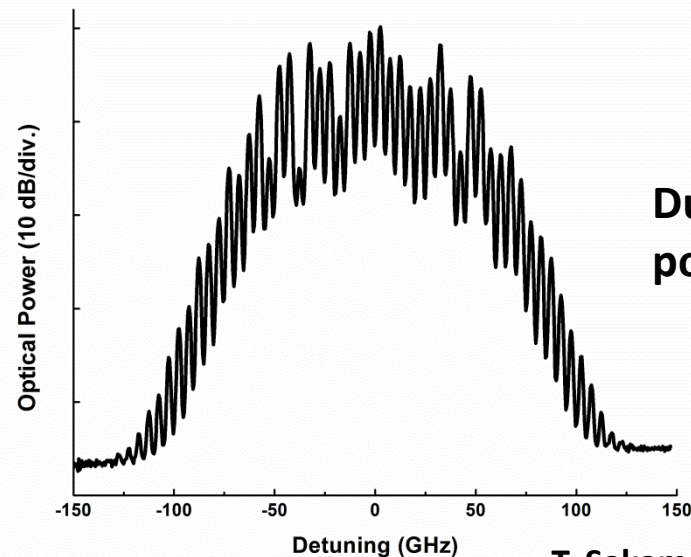
Disadvantages:

- Essentially fixed repetition rate
- Low power per tooth (nW to μ W)
- Large and expensive

An alternate approach: electro-optic modulators



Ideal for targeted measurements of selected species

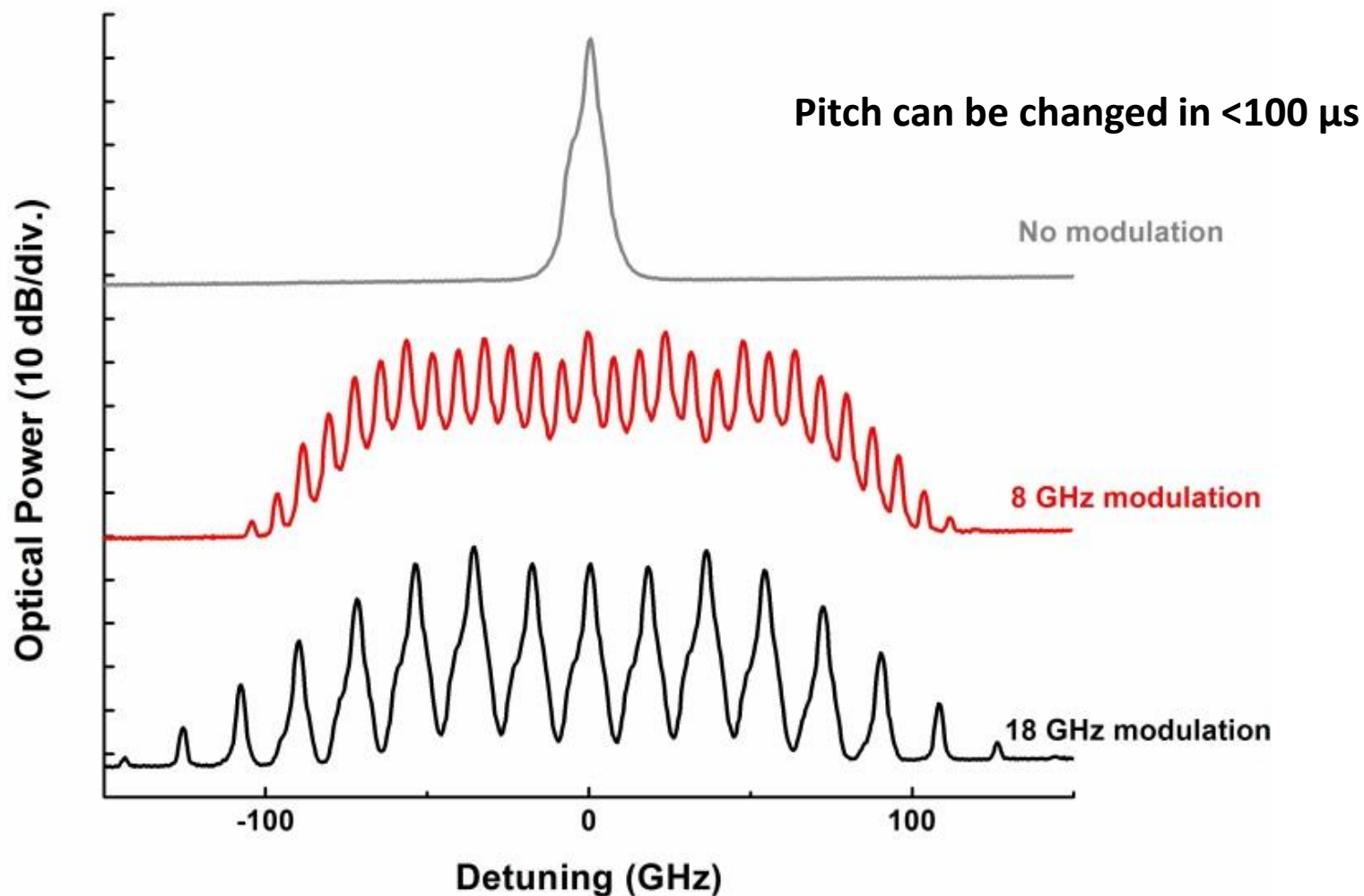


Dual-drive MZM allows for power-leveling of the comb

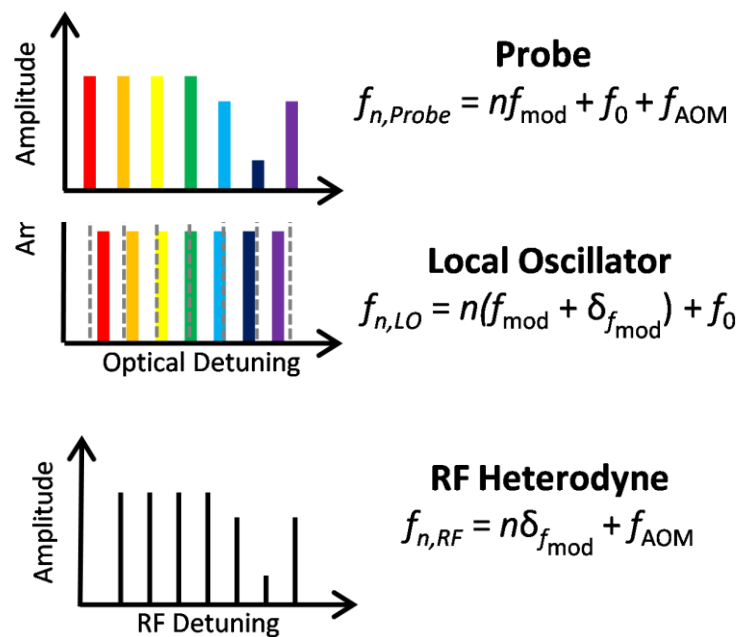
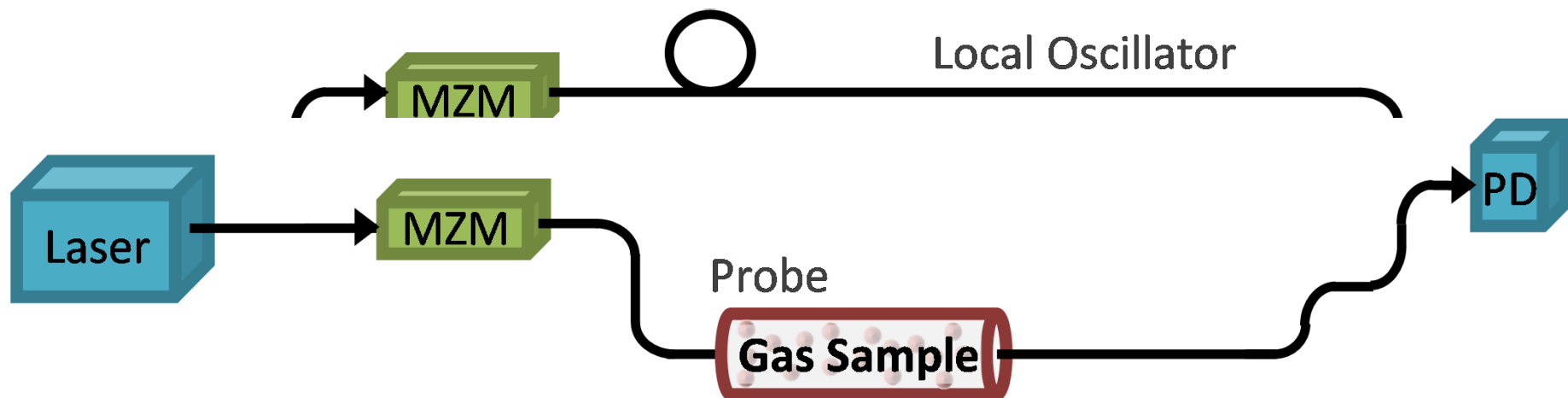
T. Sakamoto, et al., Electron. Lett., (2007).

D. A. Long, et al. Opt. Lett. (2014).

Variable pitch

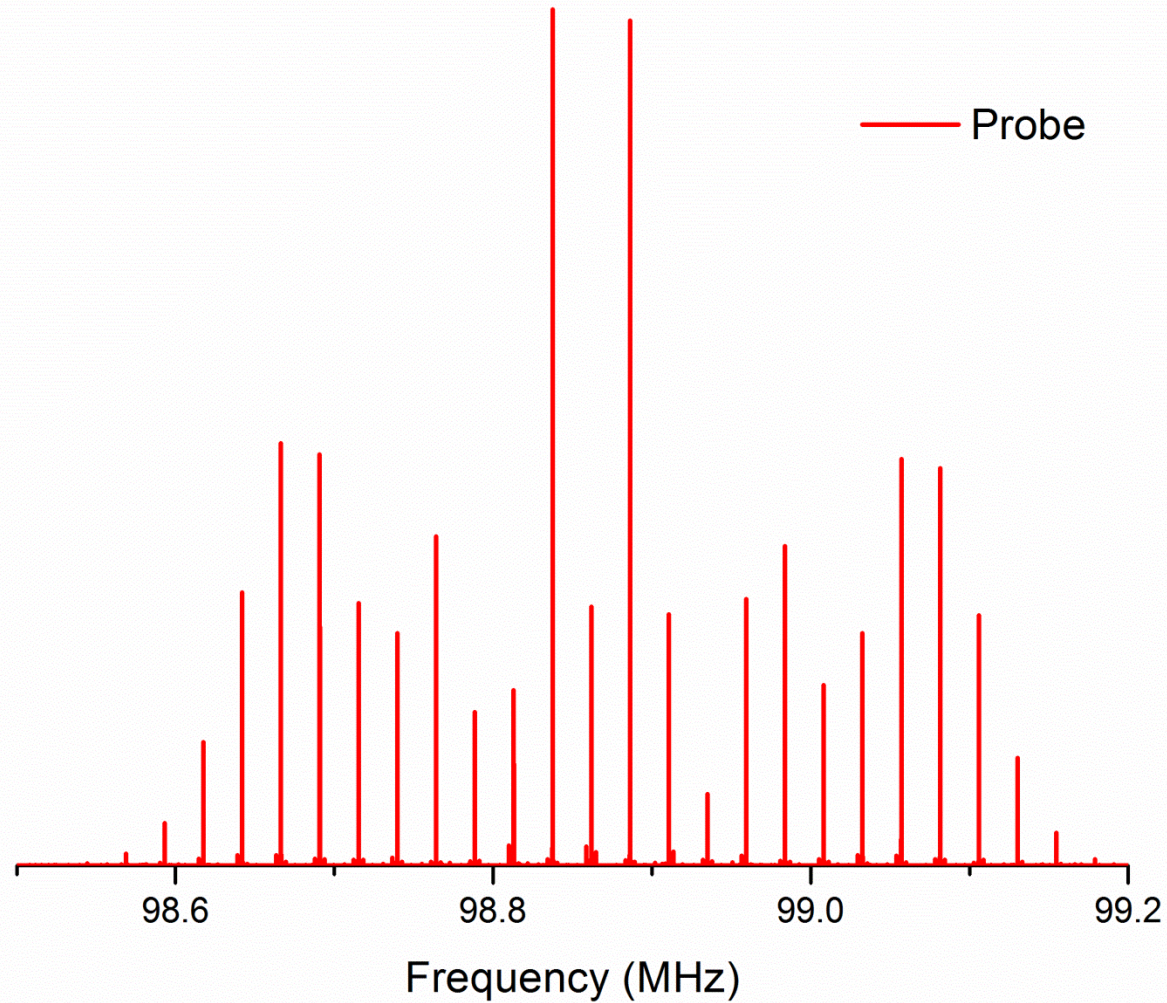


Multiheterodyne spectroscopy

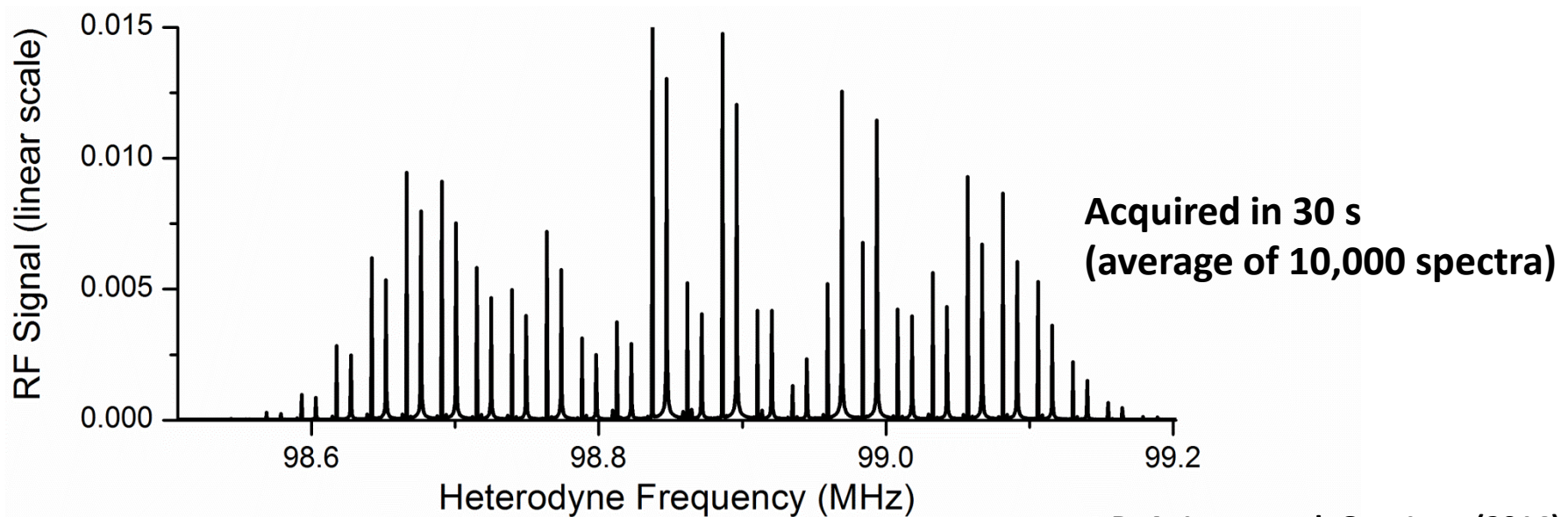


**Common mode
(no need for
phase locking)**

Referencing

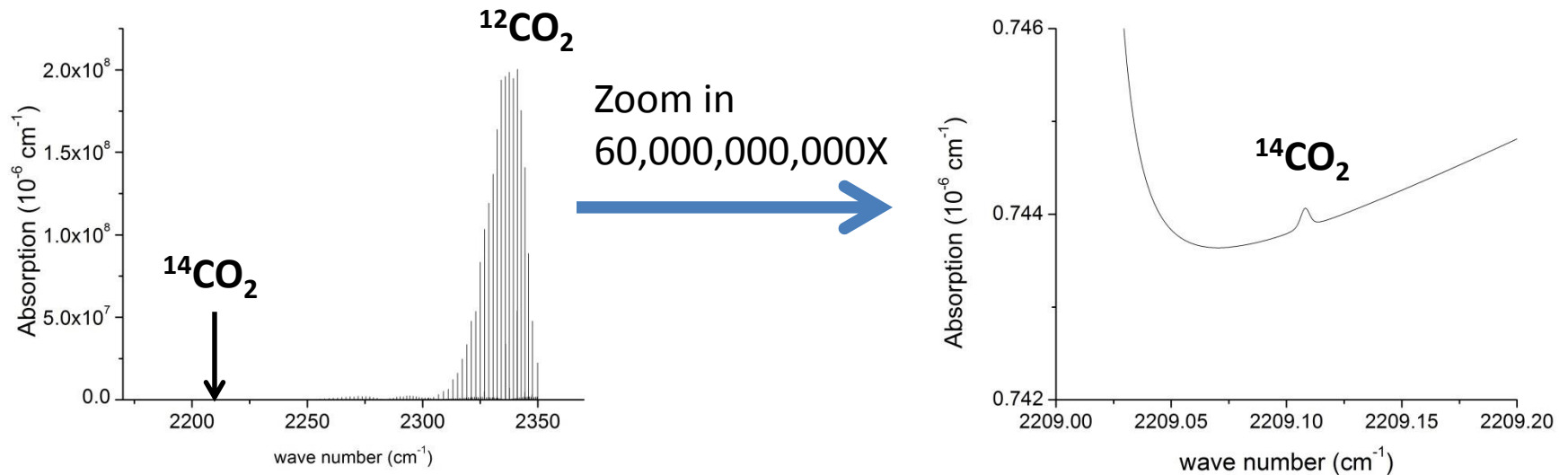


Multiheterodyne spectra



Where are we headed?

- The mid-infrared
- Probing the strongest molecular transitions allows for the lowest detection limits



Cavity ring-down spectroscopy in the mid-IR

- Instrument is up and running!

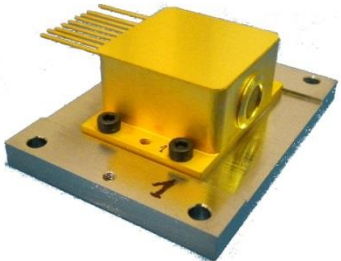
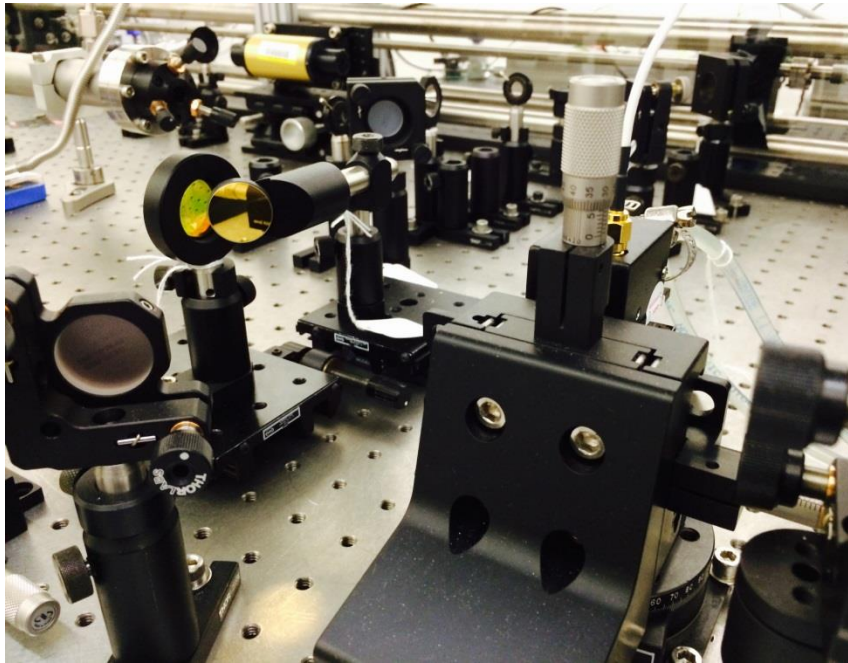
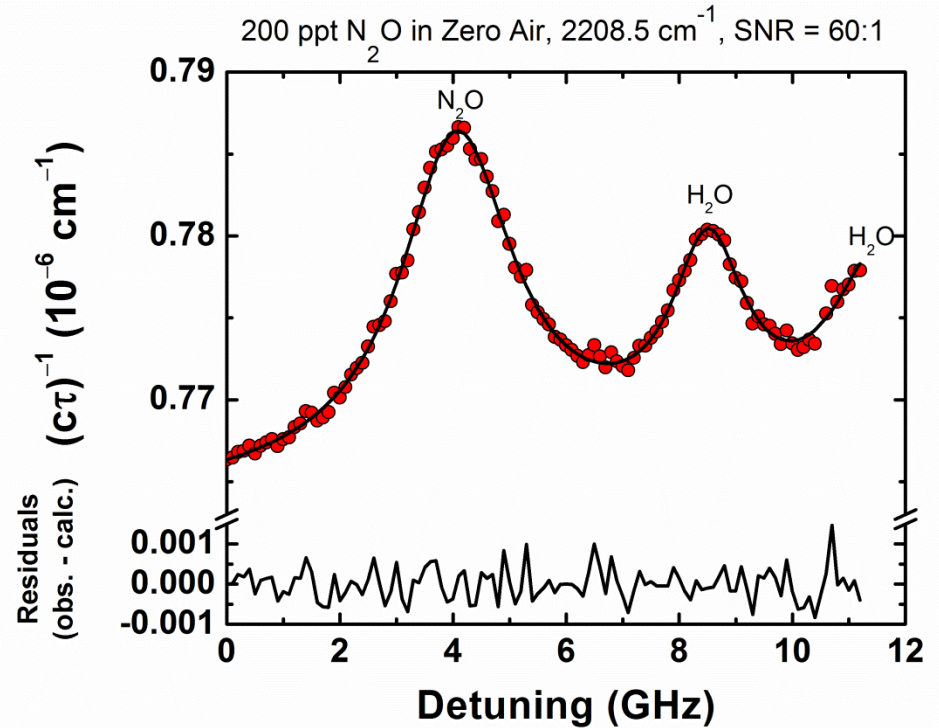


Image from Alpes Lasers



Detection limit of 3 ppt for N_2O

Conclusions

- Presented several new techniques for rapid, ultrasensitive detection of GHGs
 - **Photoacoustic spectroscopy (PAS)**
 - Relatively simply instrument allows for routine sensing
 - **Frequency-agile, rapid scanning (FARS) spectroscopy**
 - Use an EOM to step scan the laser frequency
 - Scanning rates limited only by the cavity response time
 - **Heterodyne-detected cavity ring-down spectroscopy (HD-CRDS)**
 - Make the measurement well above DC
 - Leads to quantum-noise-limited sensitivity
 - **Multiheterodyne spectroscopy with EOM-generated optical frequency combs**
 - Allows for multiplexed detection of several trace gases
 - Far lower costs and complexity than with femtosecond lasers
 - Inherently common-mode

Acknowledgements

- **Joseph Hodges, David Plusquellic, Adam Fleisher, Zachary Reed, Gar-Wing Truong, Szymon Wojtewicz, Katarzyna Bielska, Hong Lin, Qingnan Liu, Kevin Douglass, Stephen Maxwell, Roger van Zee**
 - NIST
- **NIST Greenhouse Gas Measurements and Climate Research Program**
- **NIST Innovations in Measurement Science (IMS) award**

